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Submicron ash formation from coal combustion

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Abstract

In recent years, fine particles have been found to be the cause of various harmful effects on health, and many countries have imposed restrictions on emission of these particles. Fine ash particles are formed during coal combustion in power stations and, if not collected in the air pollution control devices, are emitted into the atmosphere. The fine ash particles can remain airborne for long periods and can result in deleterious health effects when inhaled and deposited in the lungs.

Previous studies have shown that combustion of coals of different rank can result in differences in the amount and chemistry of the submicron ash particles. This study examines the variability occurring between the submicron ashes formed from coals of similar rank. Five Australian bituminous coals were burned in a laminar flow drop tube furnace in two different oxygen environments to determine the amount and composition of submicron ash formed. The experimental setup is described and the repeatability of the experiments is discussed. The variability in the submicron ash yield as a percentage of the total ash collected and the submicron ash composition are presented and discussed. This paper presents experimental results rather than a detailed discussion on its interpretation. However, the results indicate that the condensation of evaporated species is responsible for the formation of ash particles smaller than 0.3 μm.
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1. Introduction

The health effects of ambient fine particulates have been studied extensively and correlations have been observed between ambient fine particulate matter and human mortality rates [1]. Worldwide, governments acknowledge these studies and the adverse health effects of ambient fine particulates and as a result standards have been introduced to assist in reducing ambient fine particulate concentrations. In the United States, a National Ambient Air Quality Standard (NAAQS) for both 'coarse' particulate matter with an aerodynamic diameter less than 10 μm, PM$_{10}$, as well as an NAAQS for PM$_{2.5}$ is currently in effect [2]. Very recently in Australia, the National Environment Protection (Ambient Air Quality) Measure (NEPM) has been modified to include advisory reporting standards for PM$_{2.5}$, the monitoring of

which is to commence in January 2004 [3]. PM$_{10}$ has been included in the NEPM Ambient Air Quality measures since 1998 [4]. Around 80% of the electricity generated in Australia is generated by coal combustion [5], and coal-fired power generators are increasingly being required to characterise their emissions of fine particulate matter.

Studies have indicated that exposure to high concentrations of fine particulate matter may not be the sole contributor to these adverse effects, but that particle toxicology could also play an important role [6,7]. The emissions of fine ash particles and trace (toxic) elements from coal combustion are closely associated because of the relative enrichment in trace elements of these fine particles [8,9].

Although the fine particles are enriched in trace elements, their composition is not determined by these elements. It has long been established that submicron sized particles from coal combustion are primarily formed by condensation of volatilised ash, soot and char particles [10–12]. The major constituents of the submicron ash are

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the same constituents that form the bulk of the ash, however, their relative contribution varies significantly from that of the bulk ash.

In a study of the relationship between various coals and the amount and composition of the submicron ash formed, Quann and Sarofim concluded that the combustion of different coals did result in varying submicron ash yields and compositions [13]. The submicron ash yield varied between 0.54 and 9.6% of the ASTM ash, with absolute yields measured between 0.00072 and 0.0065 g of fume per gram of coal burned in a gas temperature of 1750 K and an oxygen partial pressure of 0.20 atm. In their study, the submicron ash was defined as the ash collected on the filter after the cascade impactor. The last stage of their cascade impactor had a theoretical effective cut-off diameter of 0.54 μm. In general, the submicron ash yield varied most significantly with coal rank, the lower ranked coals producing higher submicron ash yields than the higher ranked coals. Quann and Sarofim reached a similar conclusion in an earlier study [14]. In both studies it was found that for bituminous coals, the major submicron ash constituents were SiO_2, FeO, and alkali metals (Na_2O or K_2O), while the major constituents of the bulk of the ash were SiO_2, Al_2O_3, and CaO [13,14]. Both studies were conducted using the same experimental set-up, described elsewhere in more detail [15]. Flagan and Taylor have characterized small ash particles generated from the combustion of a bituminous coal in an 8–12 kW furnace [16]. They found that the main components of the ash particles less than 0.3 μm in diameter were silicon, sulphur, and sodium, all assumed present as oxides. Only a limited number of laboratory studies have sought to determine the amount and composition of submicron ash formed from the combustion of different coals. Many researchers have therefore used the model and experimental results presented by Quann as the basis for further modelling [17–19].

Combustion in enriched oxygen environments has been shown to increase the submicron ash yield [14]. In a study of the volatilisation of silica during gasification, Raask and Wilkins concluded that the amount of silica vaporized in coal-fired boilers could not be explained by direct vaporization, but had to be the result of reduction of silica to a more volatile silicon monoxide [20]. This reduction is thought to be the result of the reaction of silica with carbon monoxide to form the reduced silica monoxide and carbon dioxide [14]. A similar mechanism has been successfully applied in models to determine the extent of vaporisation of iron, aluminium, calcium and magnesium [17,18]. The modelling results have been compared with the submicron ash yield presented by Quann and Sarofim [14]. These models use char combustion models to estimate the reducing conditions and temperatures occurring on the burning char particles.

Experiments have shown that the temperature and reducing conditions in the burning char particle are important parameters for the extent of mineral matter vaporization [14]. This study examines the effect of coal selection on the submicron ash yield and composition, and the burning char particle temperature needs to be similar for each coal. Temperature measurements of burning char particles have shown that burning char particles can achieve temperatures several hundreds of degrees higher than the surrounding gas [21]. The dependence of particle temperature on char reactivity can be divided into regimes I, II, and III [22]. In regime III, the reactant gas concentration is near zero at the char surface, and the mass transfer of reactant gases through the particle boundary layer determines the char particle temperature. When char particles burn in regime III, the particle temperature is virtually independent of the char reactivity, and hence coal type [22].

In this study, a well-characterized selection of bituminous coals have been burned in air and elevated oxygen partial pressure to determine the influence of the coal characteristics and combustion environment on the submicron ash yield and its composition. The coals have been selected to represent the range of ash chemistry occurring in Australian bituminous coals. The combustion conditions have been chosen to ensure minimum variability between the attained temperature and reducing conditions during combustion of the different coals. This paper presents experimental results rather than a detailed discussion on its interpretation. Further interpretation will be provided in subsequent papers (e.g. [23]).

2. Experimental

2.1. Laboratory combustion system

The coals were burned in the Tetlow Model HTF375A drop tube furnace situated at the Chemical Engineering department at the University of Newcastle in New South Wales, Australia. The furnace is shown schematically in Fig. 1 and contains a heated vertical alumina tube of 1200 mm in length, with an effective hot zone of 1000 mm.

The coal feeder is a syringe-type feeder that pushes the coal particles into the primary gas stream. The primary gas stream entrains the coal particles and feeds them into the heated zone of the reactor via a water-cooled injector probe onto the furnace axis at a feed rate of 8 g/h. The secondary air is led through a flow straightener to provide a laminar flow profile in the hot zone of the combustor. The ratio of primary to secondary air has been chosen to ensure a laminar gas profile. Temperature controllers maintain the temperature of the hot zone of the reactor at constant temperature. The combustion products are collected through a water-cooled collection probe. To prevent condensation of combustion products on the inner shell of the collector probe, the probe is equipped with a sintered stainless steel inner shell, through which quench gas (nitrogen) is pumped. The cooled gas and combustion residue is sucked through an ash collection train, consisting of a cyclone, a seven-stage cascade impactor (MRI Model 1503), and a Teflon filter.